Pulse Self-Compression in the Subcarrier Cycle Regime

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We show that a combined medium with fast nonlinear response and plasma dispersion law can provide compression of an initially quasimonochromatic pulse below its carrier period. Solitonlike propagation for pulses with duration as short as one field oscillation is predicted. We propose that these phenomena can be observed in modern high-intensity guiding systems.

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The idea of expanding the optical pulse spectrum using self-phase modulation in a Kerr-like medium and subsequent "focusing" of the spectral components in time in a negatively dispersive medium (i.e., by means of gratings, prism pairs, or propagation effects) has proven in the past two decades to be a fundamental concept of nonlinear optics of the fs time scale, whereby a dramatic progress in many related fields has been achieved [1]. This approach enables now the generation of energetic pulses containing only two light field periods [2]. Owing to their unusual optical properties, such electromagnetic transients are of considerable interest for fundamental and applied studies. A number of propagation effects have been predicted for "single-cycle" and "half-cycle" pulses (i.e., pulse containing one or a half field oscillation) [3]. The specific behavior of weakly bound (atomic, molecular) quantum systems exposed to single and half-cycle Thz radiation has recently been reported [4].

The interest in the problem of fs-pulse self-compression is strongly motivated by the recent advances in the technique of spectral broadening of high-energy fs pulses [2]. This technique, based on pulse propagation in gas-filled hollow waveguides, provides a laser pulse guiding at an intensity level which is much higher than that achievable in optical fibers ($\sim 10^{12}$ W/cm^2 [5]). In addition, the use of prepared plasma refractive index channels for optical guiding of multiterawatt laser pulses [6], and self-channeling of an ionizing fs-pulse propagating in a gas, were recently demonstrated [7]. Because of the feasibility of combining the dielectric index of the background atoms and ions with the negative plasma electron index and the plasmalike mode dispersion of the waveguides, a phase-matched four-wave mixing and high-order harmonic generation with enormously increased conversion efficiency was recently realized [8].

The interesting feature of these systems (which has never been discussed previously) lies in the fact that the bound electron response (linear and nonlinear) of atoms and ions in the waveguide is fast and only weakly dispersive if the laser frequency is low compared to the transition frequencies (which is the case for IR pulse guiding). On the other hand, the plasma response of the combined medium shows a negative group velocity dispersion in a wide spectral range. Under certain conditions, the guiding system can be treated as a combined medium with an instantaneous nonlinear and a plasmalike linear response, therefore being an ideal object for studying pulse temporal self-action on the fs time scale.

The problem addressed here may be formulated as follows: What happens in the situation where the minimum pulse duration estimated from the conventional theory [5] of pulse self-compression,

$$\tau_{\min} \sim \tau_0 (L_{\mathrm{SPM}}/L_D)^{1/2} < T_0 = 2\pi/\omega_0 \,, \quad \text{(1a)}$$

is shorter than the incident pulse carrier period? [In Eq. (1a), $L_{\mathrm{SPM}} = 2c/(\omega_0 n_0 n_2 \varepsilon_0^2)$ and $L_D = \tau_0^2/|k_{\omega\omega}|$ are the self-phase modulation and the group velocity dispersion length of a pulse with an input amplitude ε_0 and duration τ_0.] Although the reasonable minimum duration for a quasimonochromatic wave process is limited by 1–2 field oscillations [5], condition (1a) acquires a real physical meaning if the self-compression process is treated beyond the scope of the slowly varying envelope approximation (SVEA). As we show below, it suggests generation of wave structures even shorter than the period of the incident wave. The "strong field" nature of the propagation regime can be readily seen if Eq. (1a) is rewritten in the following form:

$$\varepsilon_0 > \varepsilon_{\mathrm{cr}} = [|k_{\omega\omega}|\omega_0 c/(2\pi^2 n_0 n_2)]^{1/2}, \quad \text{(1b)}$$

showing that the extreme self-compression is expected for fields exceeding a critical field $\varepsilon_{\mathrm{cr}}$. It turns out that, for such fields, pulse nonlinear self-action in the combined medium develops on a spatial scale shorter than both the pulse dispersion length L_D and the coherence length L_{coh} for the third-order sum- and difference-frequency generation processes. These processes become phase matched in a wide spectral range and produce a spectral continuum broadened towards higher frequencies. Combined action of the blue spectral broadening and the plasmalike negative dispersion results in a formation of a temporally compressed pulse with a duration shorter than the incident pulse carrier period. We also show that the medium can support propagation of wave structures with the major part of energy confined within only one field oscillation. Because the structures are not exact soliton solutions, their integral characteristics change slowly with propagation length. Nevertheless, they exhibit a remarkable stability against perturbations and travel over distances many times exceeding their dispersive spreading length.

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Note that pulse shortening in plasma and ionized gases due to different nonlinear mechanisms has been discussed previously [9]. To our knowledge, the analysis was restricted to the SVEA approach. The mechanisms of spectral superbroadening in Kerr media are well known [5], but usually described in terms of the SVEA. By solving the exact wave equations for a pulse propagation in a dispersionless nonlinear dielectric, the important role of the sum-frequency generation process has been established in [10]. We also note that formation of subcycle structures from a "regular" pulse has been considered before for a medium of two-level atoms [11]. But the physical situation discussed in our paper has not been addressed.

To analyze pulse dynamics in the extreme regime, we introduce the following model. Consider a linearly polarized laser pulse $\varepsilon(z,t)$ propagating in the z direction in an isotropic two-component medium. The pulse evolution is described by the wave equation,

$$\partial_z^2\varepsilon - c^{-2}\partial_t^2\varepsilon = 4\pi c^{-2}\partial_t^2 P + 4\pi c^{-2}\partial_t J\,, \quad (2)$$

where the plasmalike linear component is represented by the current $J(z,t)$:

$$J(t) = \frac{\omega_p^2}{4\pi}\int_0^\infty \exp(-\nu_p\tau)\varepsilon(t-\tau)\,dt\,, \quad (3)$$

with ω_p and ν_p being the effective plasma and collisional frequency, respectively [12]. The pulse waveguiding is included by treating $\varepsilon(z,t)$ as an on-axis value of the corresponding spatial mode; ω_p then accounts for both the plasma electron response and the plasmalike mode dispersion law, and ν_p can be interpreted in terms of frequency dependent mode attenuation [8].

The transition frequencies $(E_j - E_k)/\hbar$ of the atomic (ionic) component are assumed to lie well above the laser field frequency ω. This means that dispersion effects are small, and the response is determined by the instantaneous value of the field $\varepsilon(t)$:

$$P(t) = \chi_0^{(1)}\varepsilon(t) + \chi_0^{(3)}\varepsilon^3(t)\,. \quad (4)$$

Here $\chi_0^{(1)}$ and $\chi_0^{(3)}$ are the medium linear and nonlinear polarizability in the static field limit where they are positively signed [13], and $\chi_0^{(3)} \sim (\mu_a/\hbar\omega_a)^2\chi_0^{(1)}$ (μ_a and ω_a being the characteristic dipole moment and transition frequency of the system).

In terms of the medium dispersion $k(\omega) = (\omega/c) \times [1 + 4\pi\chi_0^{(1)} - (\omega_p/\omega)^2]^{1/2}$ (assuming $\nu_p \ll \omega$) and the Kerr constant $n_2 = 3\pi\chi_0^{(3)}/n_0$ with $n_0 = [1 + 4\pi\chi_0^{(1)}]^{1/2}$, the critical field can be rewritten as $\varepsilon_{\rm cr} = (\omega_p/\omega_0)[6\pi^3\chi_0^{(3)}]^{-1/2}$. We also introduce the unit propagation length $L_0 = (2\pi n_0)^2 c\omega_0/\omega_p^2$ associated formally with the spreading length of a one-oscillation-long ($\tau_0 = T_0$) pulse in the plasma. The dispersion length of a pulse with $S = (\tau_0/T_0)$ field oscillations is then $L_D = L_0 S^2$.

In the first series of numerical studies of the full Eqs. (2)–(4), we analyzed nonlinear propagation of long input pulses with $S \gg 1$. The input field envelope was assumed to be sech-shaped with an amplitude value ε_0 and duration τ_0. As far as the condition $\varepsilon_0 \ll \varepsilon_{\rm cr}$ was satisfied, all the specific features of the N-soliton solution [5] of the nonlinear Schrödinger equation (NLSE) were observed. For $N \gg 1$, a self-compression with a factor $\sim N$ occurred.

The picture of pulse evolution changes dramatically for input amplitudes $\varepsilon_0 \geq \varepsilon_{\rm cr}$. In Fig. 1 is shown the evolution of the pulse intensity [1(a)] and the field spectrum [1(b)] with the propagation distance for an input pulse corresponding to 2.5 solitons of the NLSE (the intensity dynamics is presented to demonstrate more clearly the compression effect). As can be seen, in the course of propagation, the initially symmetrical pulse experiences a progressing asymmetry with a flatter leading edge and a steeper trailing edge. The pulse spectrum exhibits a well-pronounced asymmetry too, with a long blue wing extending to frequencies above 3ω. The contribution of sum-frequency generation to the spectral superbroadening is evident already at the initial stage in the frequency region near 3ω [Fig. 1(b), $z = 2L_0$]. As a result of the combined action of the continuum generation and the plasmalike dispersion, a very short electromagnetic substructure is formed at the trailing edge of the pulse [Fig. 1(a), $z = 6L_0$]. The intensity of the subpulse is nearly 3 times higher than that of the input pulse, while its duration is even shorter than the input pulse carrier period $T_0 = 2\pi/\omega_0$, in

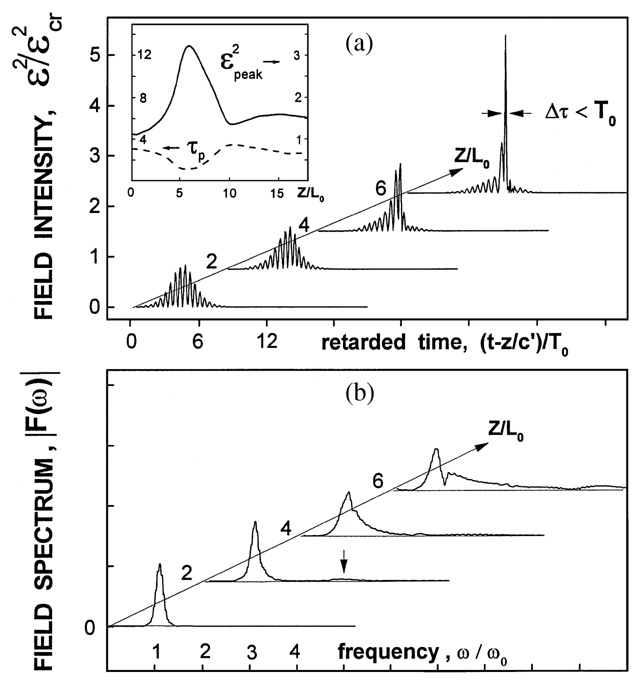

FIG. 1. Self-compression of a laser pulse in a medium with a fast nonlinear and plasmalike linear response: (a) the field intensity, (b) the field spectrum at different propagation lengths Z. ε^2 is normalized by $\varepsilon_{\rm cr}^2$, Z in units of L_0. The inset shows the pulse peak intensity and effective duration (τ_p) with 70% of the full energy; τ_p is in units of the input carrier period T_0; $\omega_p/\omega_0 = 5 \times 10^{-2}$; $\nu_p/\omega_0 = 5 \times 10^{-4}$. The buildup of the third harmonic signal is shown with an arrow.

agreement with the prediction of relation (1a). The energy within the peak is about one third of the pulse full energy. The compression effect is evident from the inset of Fig. 1 showing variation with z of the pulse effective duration (with 70% of its full energy) and the peak intensity. It was found that for higher ε_0 even shorter subcycle wave forms can be produced, but the efficiency of the process (the relative part of the energy within the peak) decreases, so that the optimum field lies in the region $\varepsilon_0 = (1-2)\varepsilon_{\mathrm{cr}}$.

The pulse propagation differs qualitatively for $\varepsilon_0 \gg \varepsilon_{\mathrm{cr}}$. Rather than a smooth spectral distribution such as that in Fig. 1(b), a generation of separated superbroadened lines corresponding to odd harmonics of the field occurs. In this case, no temporal compression is observed.

The above features of the self-compression can be better understood if one analyzes the spectral changes at the initial stage where the nonlinear contribution dominates over the linear plasmalike response. By neglecting the plasma term $(\partial J/\partial t)$ in the right-hand side of Eq. (2) compared to the bound electron term $(\partial^2 P/\partial t^2)$, the wave equation can be solved exactly to obtain solution in the implicit form:

$$\varepsilon(z,t) = F[t \mp (z/c)\sqrt{\kappa(\varepsilon)}\,], \tag{5}$$

where $\kappa(\varepsilon) = \partial(\varepsilon + 4\pi P)/\partial\varepsilon = \partial_\varepsilon\{\varepsilon + 4\pi(\chi^{(1)}\varepsilon + \chi^{(3)}\varepsilon^3)\}$, and $F(\xi)$ is the field temporal shape at $z = 0$. Solution (5) describes a traveling (in $\pm z$ direction) electromagnetic wave with the time-dependent velocity $v = c/\sqrt{\kappa(\varepsilon)}$ determined by the value of the field at a given moment. As is well known from the literature [14], the regime leads to the shock wave generation. Assuming the input field in Eq. (5) in the form $F(t) = \Psi(t)\sin(\omega t)$, where $\Psi(t)$ is the envelope function, and considering the field distortions in the lowest order we have:

$$\begin{aligned}\varepsilon(z,\tau) = {}& \Psi(z,\tau)\sin[\omega\tau - \tfrac{1}{2}z\omega\alpha\Psi^2(z,\tau)] \\ &+ \tfrac{1}{2}z\alpha\omega\Psi^3(z,\tau)\cos[3\omega\tau + \tfrac{3}{4}z\omega\alpha\Psi^2(z,\tau)],\end{aligned} \tag{6}$$

where $\tau = (t - z/c')$ with $c' = c/n_0$, $\alpha = (6\pi c'/c^2)\chi_0^{(3)}$, and the $\Psi(z,\tau) = \Psi(\tau - z\alpha\Psi^2)$ depends on the variables implicitly. According to Eq. (6), an initially smooth envelope $\Psi(\tau)$ evolves towards flattening of its leading and steepening of its trailing edge, since the points where the field intensity is higher move with smaller velocity: $v = c'/(1 + \alpha c'\Psi^2)$. This leads to a formation at the pulse trailing edge of a structure close to the envelope shock wave [5] that is seen in Fig. 1(a), at $z/L_0 = 2$. The spectral broadening can roughly be treated as a nonlinear frequency modulation of the fundamental $(\partial/\partial t)\phi_\omega = (\partial/\partial t)(-\frac{1}{2}z\omega\alpha\Psi^2)$ in the presence of steepening of the pulse envelope. Together with the competing third harmonic generation process, it forms the asymmetric blue wing. Because the positive frequency modulation is produced in the region of the envelope shock formation, temporal compression by the plasma dispersion is observed at the pulse trailing edge. Estimating from (6) the third harmonic generation length by $L_{3\omega} \approx 2c^2/(3\pi c'\omega\chi_0^{(3)}\varepsilon_0^2)$ and calculating the wave number mismatch, $\Delta k = 3k(\omega) - k(3\omega) = (\frac{8}{3})\omega^2 k_{\omega\omega}$, we find that, for the $\varepsilon_0 > \varepsilon_{\mathrm{cr}}$ condition, $L_{3\omega} < L_{\mathrm{coh}} = \pi/\Delta k$ becomes fulfilled supporting the above considerations. For even higher incident fields $\varepsilon_0 \gg \varepsilon_{\mathrm{cr}}$, phase matching is realized for multiple harmonics, and irregularly broadened spectra are observed.

The self-compression dynamics raises the interesting question of the existence of solitonlike propagation in the region of extremely short durations. We have found numerically that the wave forms with the major part of the field energy concentrated within one electromagnetic cycle ($s = 1$) indeed can propagate in the regime where the continuum generation process is balanced by the plasmalike dispersion. Figure 2 displays the evolution of the field [2(a)] and spectrum [2(b)] of such a pulse. The "carrier" substructure of the pulse propagates with a velocity slightly exceeding its group velocity which leads to the specific oscillating behavior. The pulse as a whole demonstrates remarkable stability at propagation distances many times longer than the pulse dispersive spreading length L_0. To illustrate this, in Fig. 2, superimposed on the field at $z = 24L_0$, is also shown the field of the same input pulse propagated to this distance in the linear regime ($\chi_0^{(3)} = 0$). Since the nonlinearity (4) in the solitonlike pulse is not ideally matched by the plasma dispersion, the pulse irradiates a high- and low-frequency field, but the corresponding

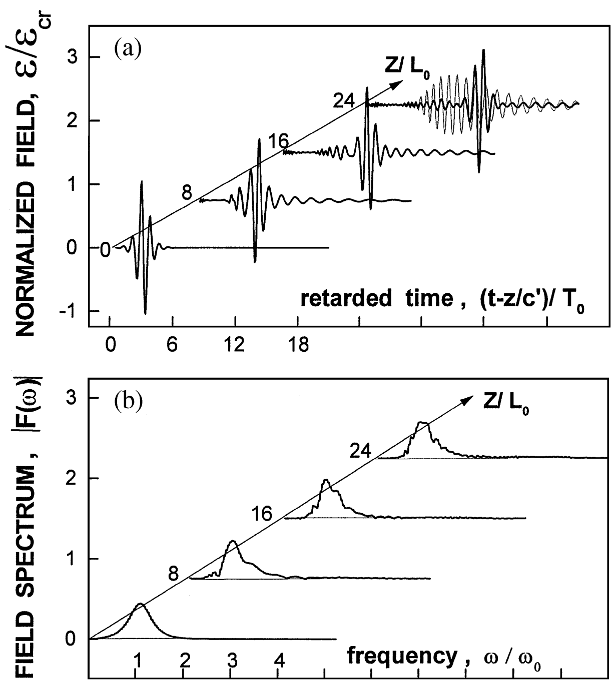

FIG. 2. The solitonlike dynamics of a pulse carrying 70% of its energy within one field oscillation: (a) the pulse electric field, (b) the pulse spectrum at different propagation lengths Z. Superimposed on the pulse field, at $Z = 24L_0$, is also shown the field of the same input pulse propagated to this distance in the linear regime. Normalization units as in Fig. 1.

energy losses are only few percents. [Note that in the case $\chi_0^{(3)} > 0$ of interest soliton effects exist for a medium dispersion law $k(\omega) = \omega + \beta\omega^3$, $\beta < 0$ [15].]

The effect of perturbations on the quasisoliton dynamics was examined by varying the pulse input amplitude (Fig. 3). For stronger input pulses, we observed an increasing spectral broadening leading, after some transition stage, to a formation of rather stable shorter pulses with the similar temporal structure and the center of gravity of the spectrum shifted to higher frequencies. In contrast, weaker input pulses evolved towards the formation of stable red-shifted pulses with a narrower spectrum and a duration containing more than one oscillation.

By setting the characteristic self-action length of the solitonlike pulse $L_{NL} \sim \tau_0 c/(2\pi\chi_0^{(3)}\varepsilon_0^2)$ equal to its spreading length in the plasma component $\sim L_0 = (2\pi)^3 c/(\omega_p^2 \tau_0)$, we find the relation between the pulse duration and field strength: $\tau_0 \approx 2\pi(2\pi/\omega_p) \times (\chi_0^{(3)})^{1/2}\varepsilon_0$, showing that shorter pulses have weaker field strengths. This can be explained by the fact that the plasma dispersive action generally decreases with increase of field frequency.

Let us make some estimations. Consider pulse guiding in a plasma channel created in a noble gas. The plasmalike contribution to the waveguide dispersion, $-(\omega_p/\omega)^2$, dominates if the frequency-dependent part $[\chi^{(1)}(\omega) - \chi_0^{(1)}]$ of the atomic (ionic) linear index is small. Expanding the bound electron polarizability in a series with the small parameter $(\omega/\omega_{kg}) \ll 1$ [where ω_{kg} is the transition frequency between the ground (g) and the excited state (k) of the atom or ion], the requirement can be written in the following form:

$$(8\pi/\hbar)\sum_{\sigma,k} N^{(\sigma)}(|\mu_{kg}^{(\sigma)}|^2/\omega_{kg}^{(\sigma)})\,(\omega/\omega_{kg}^{(\sigma)})^2 \ll (\omega_p/\omega)^2, \tag{7}$$

where index k numbers the bound excited states with the dipole moment μ_{kg}; $N^{(\sigma)}$ is the concentration of the atomic $(\sigma = a)$ and ionic $(\sigma = i)$ component. For instance, for a laser photon $\hbar\omega_0 \approx 1$ eV, $\hbar\omega_{kg} \sim 10$ eV, $\mu_{kg} \sim 10^{-18}$ esu, and the plasma channel radius 50 μm, condition (7) can be satisfied if $N_e \sim N^{(i)} \sim 10^{18}$ cm^{-3}, $N^{(a)} \sim 3 \times 10^{19}$ cm^{-3}. Assuming $n_2 \sim 10^{-17}$ esu ($\tilde{n}_2 = 10^{-19}$ W/cm^2 [2]), the critical field is calculated to be $\approx 2 \times 10^7$ V/cm ($I_{\rm cr} \sim 10^{14}$ W/cm^2). For a pulse with $\tau_0 = 10$ fs, self-compression down to one oscillation (3 fs) occurs at $L \approx 2$ cm. The estimated medium ionization [16] is still small compared to the prepared electron concentration.

In conclusion, we have shown that a combined medium with fast nonlinear response and plasmalike dispersion offers interesting possibilities to observe nonlinear effects on the sub-fs time scale. Pulse self-compression below the carrier period and solitonlike propagation of pulses with energy contained within one field oscillation are predicted. Practically, our analysis shows that such phenomena can be realized in modern high-intensity guiding systems.

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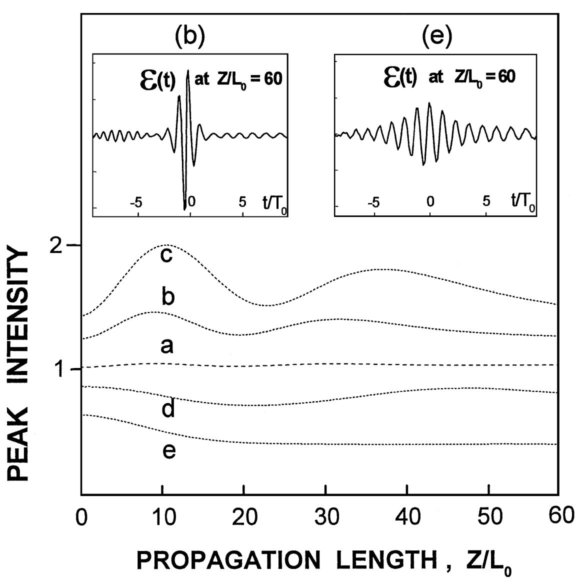

FIG. 3. The effect of perturbations on the solitonlike propagation: (*a*) the peak intensity of the unperturbed solitonlike pulse and [(*b*),(*c*),(*d*),(*e*)] of pulses with the same input form but with perturbed amplitudes. The insets show the temporal structure of the fields formed at $Z = 60L_0$ for the perturbed pulses (*b*) and (*e*).

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